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Influence of demixing effect on the temperature in wall-stabilized SF_6 arcs

J J Gonzalez, J B Belhaouari and A Gleizes

CPAT, URA CNRS 277, Université Paul Sabatier, 118 route de Narbonne, F31062 Toulouse Cédex, France

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Abstract. The demixing effect in a SF_6 wall-stabilized arc leads to sulphur depletion in the central part of the arc where the ratio between fluorine concentration and sulphur concentration becomes higher than six. We have calculated material functions for different real or fictitious gases SF_M, with $6 \leq M \leq 15$ (specific heat, electrical and thermal conductivities, net emission coefficient) and used them in a one-dimensional model based on Patankar's algorithms. The electric field and the temperature radial profile have been calculated versus the current intensity using 5 mm and 3.2 mm arc diameters. Comparison with experimental results in the literature shows that the influence of demixing on the axis temperature is greater than the experimental error. Thus, we conclude that validating material functions of SF_6 plasma by comparing experimental and theoretical values of temperature in wall-stabilized arcs needs to take into account demixing effects.

1. Introduction

In recent years, plasma modelling has developed greatly in terms of performance and to make predictions. Most industrial plasmas are derived from mixtures of gases; the large number of species present in such plasmas and the occurrence of demixing make their experimental characterization very difficult. In most cases, local thermodynamic equilibrium (LTE) is assumed to exist. Experimental analyses of wall-stabilized SF_6 arcs have shown that demixing effects change the chemical composition of the plasma (Schulz-Gulde 1980, Vacquié *et al* 1985, Razafinimanana *et al* 1993), and that demixing can have a large influence on macroscopic arc parameters (Murphy 1995).

The phenomenon of demixing can be explained simply if we consider an arc discharge in a molecular gas composed of two species of atoms with different ionization potentials. The neutral atoms diffuse from the edges towards the axis of the arc and the ions diffuse with the electrons from the axis towards the walls. The neutral atoms of the species with the lowest ionization potential will, statistically, become ionized before the others and diffuse towards the walls in the ambipolar flux. This leads to a lowering of the concentration of this species with respect to equilibrium near the axis. In the case of a SF_6 arc the low-potential species is sulphur so that the ratio M (called here the demixing factor) of the fluorine concentration (atom + ions) on the sulphur concentration is higher than six near the axis.

In spite of the existence of demixing effects, the calculated values of transport coefficients for SF_6 plasma, are sometimes validated by comparing experimental values of temperature (from a wall-stabilized arc) with values computed by a model based on the use of the transport coefficients. In this paper we first present the calculation of the material functions (transport coefficients and radiation losses) of SF_M plasma for different values of M ($M \geq 6$) and, second, the calculation of temperature in these kinds of mixture, for different values of radius, and current intensity. The comparison between our computed temperature values with experimental values in the literature will allow study of the influence of demixing on the temperature of SF_6 wall-stabilized arcs and evaluation of the validity of the material functions of SF_6 plasmas.

2. Material functions

Two types of material functions are required for the model—transport coefficients, such as electrical and thermal conductivity, and radiation.

The first functions have been computed using the classical method of Chapman and Enskog already used for SF_6 and SF_6–Cu (Chervy *et al* 1994). This method considered an ideal plasma in LTE. All the compounds were taken to be gaseous. Associating the equations of Saha, Gulberg–Waage and Dalton with those of electrical neutrality and conservation of stoichiometric equilibrium enables the particle number densities to be calculated as a function of temperature, pressure and stoichiometric ratio.

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The pressure corrections and the lowering of the ionization potential were taken into account. The internal partition functions of each species were determined differently according to their structure. The mass density was directly obtained from the composition mixture.

The theoretical study of transport coefficients is based on the resolution of Boltzmann's integro differential equation using the method of Chapman–Enskog. Using the third-order approximation of this method, the electrical conductivity expression represents the electron component of electrical conductivity which neglects the contribution of the current due to ions. The thermal conductivity is equal to the sum of several components, i.e. the translation thermal conductivity, the internal conductivity and the reaction thermal conductivity. In the transport coefficient expressions, special functions called collision integrals were introduced. Their calculations differ according to the type of interaction considered and we used different methods given by Chervy *et al* (1994). All expressions and all data (reaction energies, molecular constants, interaction potentials, radii, etc) used for these calculations are reported by Chervy *et al* (1994).

Radiative energy loss was calculated using the net emission coefficient assuming an isothermal and homogeneous plasma of radius R_p. The method, described by Gleizes *et al* (1993), takes into account the emission and absoprtion of radiation due to continuum and lines. Self-absorption of each line is calculated by means of an escape factor. It has been previously shown that the use of a net emission coefficient in wall-stabilized arc models is a good approximation of the radiative losses.

Figure 1 shows the electrical conductivity versus temperature for three values of M $(6, 10, 15)$. Under 15 000 K, the electrons are provided mainly from the ionization of sulphur. Then, if M increases, the sulphur and electron populations decrease and thus the electrical conductivity decreases. In figure 2 we have plotted for the same gases the thermal conductivity versus temperature. For the thermal conductivity of SF_6, we find three peaks centred at around 1800, 2200 and 2800 K corresponding to the dissociations of SF_6, SF_4 and SF_2 respectively. When M increases, the amplitude of these peaks diminishes which is due to the reduction of the number densities of these species with the stoichiometric proportion of S atoms, and we note the occurrence of F_2 at temperatures lower than 2000 K which is due to the conservation of the new stoichiometric equilibrium. The dissociation of F_2 gives rise to a minor peak around 1300 K.

We can see in figure 3 ($R_p = 2$ mm) that, as M increases, the relative proportion of sulphur decreases and then the net emission coefficient decreases because radiation emitted by sulphur is much more important than that from fluorine species. The material functions are plotted for temperatures up to 18 000 K, which is higher than the maximum axis temperature measured in wall-stabilized arcs.

3. Modelling

Our purpose here is not to simulate very precisely the complicated phenomena leading to the demixing effect, but to quantify the influence of demixing on the temperature. The main assumptions of the model are as follows.

(i) The plasma is in LTE with a cylindrical symmetry.
(ii) The electric field is constant and uniform.

These two hypotheses are generally admitted in a wall-stabilized arc (Lowke and Mitchell 1983, Gleizes *et al* 1987). The first has been experimentally validated (Vacquié *et al* 1982) whereas the second is directly deduced from the fact that the plasma properties are independent of the axial coordinate.

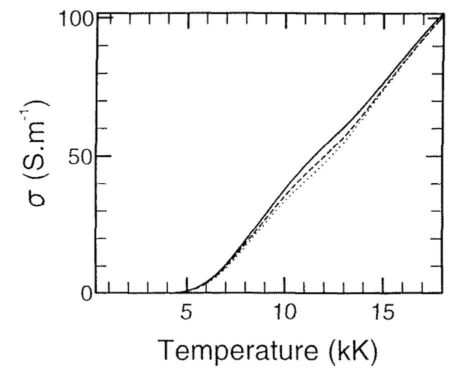

Figure 1. Electrical conductivity versus temperature: SF_6, ——; SF_{10}, – – –; SF_{15}, · · · · · ·.

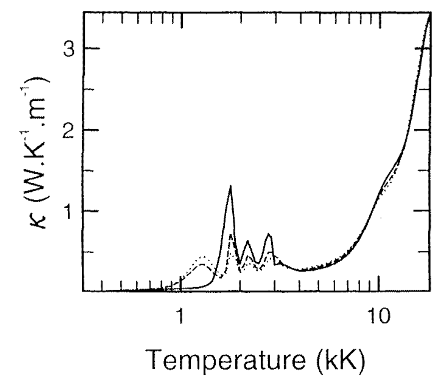

Figure 2. Thermal conductivity versus temperature: SF_6, ——; SF_{10}, – – –; SF_{15}, · · · · · ·.

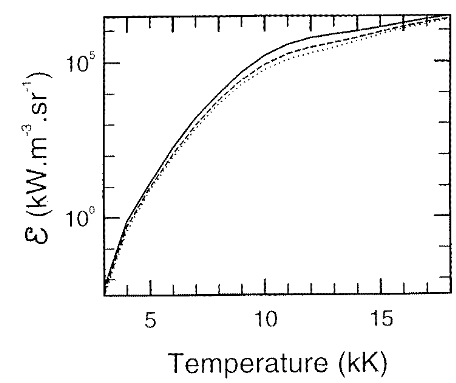

Figure 3. Net emission coefficient versus temperature: SF_6, ——; SF_{10}, – – –; SF_{15}, · · · · · ·.

(iii) The pressure is uniform and there is no flow (convection).

Calculations made in a two-dimensional wall-stabilized arc by Lowke (1979) suggests that radial pressure gradients might be neglected. The hypotheses concerning axial and radial convection have been analysed and justified theoretically and experimentally in the case of a wall-stabilized arc by Gleizes (1981).

(iv) For the radiation we use the net emission coefficient.

The assumption that radiation losses can be represented by a net emission coefficient, giving power loss in W m^{-3} as a function only of temperature, enables reasonably accurate estimates of central-arc temperature even if there is self-absorption of radiation (Lowke 1974). So in the central part of the arc, radiation energy losses are considered with a net emission coefficient (Gleizes *et al* 1993) R_p being dependent on the actual arc diameter ϕ : $R_p = 2$ mm when $\phi = 5$ mm and $R_p = 1$ mm when $\phi = 3.2$ mm. In the outer part of the plasma, absorption of radiation is taken into account by a crude approximation deduced from the work of Lowke and Mitchell (1983). We consider that a part of the radiation emitted from the hottest regions is radially absorbed in the regions where the temperature is lower than a critical value. In our work we have chosen this critical value to be 10 000 K. Lowke and Mitchell (1983) assumed that all the emitted radiation was absorbed in the warm or cold plasma and gas. From the experimental and theoretical work of Gleizes *et al* (1995) we assume that only 2/3 of the emitted radiation is absorbed. The net emission coefficient ϵ_N is calculated (Gleizes *et al* 1993) to be appropriate for the centre of an isothermal cylindrical plasma of a given radius R_p. We calculate the radiation flux density F_{rad} using the relation:

$$F_{rad}(r) = \frac{1}{r}\int_0^r 4\pi\epsilon_N r' \mathrm{d}r'. \tag{1}$$

At the edge of the arc, ultraviolet radiation from the arc centre is absorbed. We account for this effect by making ϵ_N negative when the temperature is lower than 10 000 K and considering a homogeneous distribution where a fraction equal to 2/3 of F_{rad} will be absorbed. Only 1/3 of the radiation emitted by the arc escapes the plasma.

(v) In order to have an approximation of the demixing effect, we consider that in the central part of the arc, the factor M is constant (and equal to 10 or 15, for example), whereas in the outer part we consider stoichiometric equilibrium ($M = 6$). The limit between the two regions corresponds to the position of the isotherm $T = 9500$ K. In practical calculation of this study we use for all the results the material functions of SF_6 for $T < 9500$ K and those of SF_M ($M > 6$) for $T > 9500$. In experimental situations, M depends on various parameters such as the arc diameter, the current intensity and the radial position (Razafinimanana *et al* 1993). From previous measurements given in the literature (Schulz-Gulde and Worzyk 1983, Vacquié *et al* 1982, Gleizes *et al* 1987), we can consider that the axis values of M are roughly between 10 and 20 for an arc diameter between 3 and 5 mm, when the current is of the order of several tens of amperes.

The temperature can be calculated by the energy balance equation coupled with Ohm's law. In cylindrical coordinates, the energy balance equation may be written as

$$\sigma E^2 - u + \frac{1}{r}\frac{\partial}{\partial r}\left(r\kappa\frac{\partial T}{\partial r}\right) = 0 \tag{2}$$

where σ and κ are respectively the electrical and the thermal conductivity, u is the radiation term and T the temperature. Ohm's law states

$$E = \frac{I}{G} = \frac{I}{2\pi\int_0^R \sigma r \mathrm{d}r} \tag{3}$$

where E is the electric field, I the current intensity and G the conductance.

For this study the number of radial points leads to a radial step $\Delta r = 2.5$ μm with a constant step. The resolution is based on the algorithms of Patankar (1980). The boundary conditions are $(\partial T/\partial r)_{r=0} = 0$ and $T_w = 300$ K, T_w being the wall temperature (at $r = R$). The $T(r)$ profile is computed starting from an arbitrary initial parabolic temperature profile.

4. Results

The results are presented for two arc diameters $\phi = 5$ mm and $\phi = 3.2$ mm. The variations of the axis temperature versus the current intensity for an arc diameter of 5 mm are plotted in figure 4. The calculated curves obtained for $M = 6$ (SF_6), $M = 10$ (SF_{10}) and $M = 15$ (SF_{15}) are compared with experimental results reported by Schulz-Gulde and Wotzyk (1983) and Vacquié *et al* (1982). Several comments can be deduced from the results of figure 4.

(i) An increase of the current intensity leads to a trivial result, i.e. an increase of the axis temperature.

(ii) The values calculated without taking demixing into account ($M = 6$) are not in agreement with the experimental results. The experimental values are systematically higher than those calculated ($M = 6$) and the difference (about 1000 K when $I > 60$ A) is larger than the experimental uncertainty.

(iii) We observe good agreement between experimental and theoretical values of the axis temperature when the demixing effect is included in the calculation. Furthermore, comparison between all the results shows that the demixing effect should be moderated for low current intensity (good agreement between experience and theory for $I < 150$ A and $M = 10$) and should increase with the current. This is in good agreement with experimental values of the factor M given by Schulz-Gulde (1980) and Gleizes *et al* (1987); radial gradients increase with the current leading to an important demixing process.

The fact that the demixing effect leads to an increase of the axis temperature is mainly due to two phenomena. The first one is the decrease of net emission from the hottest regions when M increases, which corresponds to a decrease of energy loss and thus to an enhancement of temperature. The second is the decrease of the electrical conductivity when demixing is accounted for—at a given

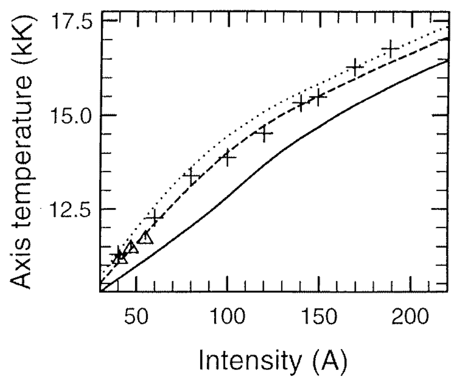

Figure 4. Variation of the axis temperature versus the current intensity for an arc diameter of 5 mm—comparison of experimental and theoretical results: Schulz-Gulde and Worzyk (1983), +; Vacquié *et al* (1982), △; SF_6, ——; SF_{10}, – – –; SF_{15}, · · · · · ·.

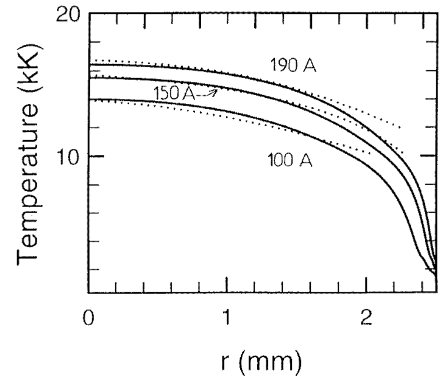

Figure 6. Comparison of experimental (· · · · · ·) and theoretical (——) radial temperature profile obtained in SF_{10} with different current values. Upper curves, 190 A; central curves, 150 A; lower curves, 100 A.

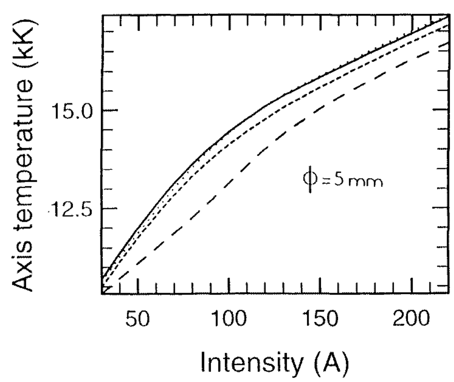

Figure 5. Variation of the axis temperature versus the current intensity for an arc diameter of 5 mm. SF_{15}, ——; results with κ of SF_6 and the other functions of SF_{15}, · · · · · ·; results with σ of SF_6 and the other functions of SF_{15}, - - - -; results with ε of SF_6 and the other functions of SF_{15} — — —.

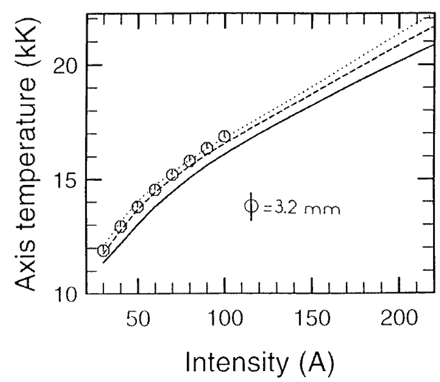

Figure 7. Axis temperature versus current intensity for an arc diameter of 3.2 mm (experimental and theoretical values). Schulz-Gulde and Worzyk (1983) $T(0) = 4455\, I^{.289}$ for 30 A $\leq I \leq$ 100 A, ○; SF_6, ——; SF_{10}, – – –; SF_{15}, · · · · · ·.

current, the decrease of the electron number density due to demixing must be compensated by a higher temperature. In order to determine which is the most important mechanism, we have plotted in figure 5 four theoretical curves of axis temperature, computed with $M = 15$ corresponding to: the results shown in figure 4; the results obtained by considering in the central part the electrical conductivity of SF_6 and other material functions of SF_{15}; results with the net emission coefficient of SF_6 and other functions of SF_{15}; results with the thermal conductivity of SF_6 and other functions of SF_{15}. These curves show that the material property most influenced by demixing is radiation. The influence of the variation of thermal conductivity at high temperature due to demixing, is very low.

Good agreement between experimental and theoretical (obtained with $M = 10$) values is also shown in figure 6 where temperature profiles for three current intensities and $\phi = 5$ mm are plotted. For the highest intensity the experimental axis temperature is slightly higher than that calculated because demixing is probably stronger than for $M = 10$. On the other hand the difference in temperature profile near the wall may be due to departures from LTE.

Figure 7 shows the same kind of results as in figure 4, but for an arc diameter $\phi = 3.2$ mm, the experimental results shown being those of Schulz-Gulde and Worzyk (1983). We observe the same tendencies as in figure 4, i.e. good agreement between experiment and theory when demixing is taken into account in the calculation. We note that for the same current intensity, the demixing effect is stronger for $\phi = 3.2$ mm than for $\phi = 5$ mm. This is due to the strong temperature and number density radial gradients leading to an important ambipolar diffusion favouring the demixing process.

For arc diameters of 3.2 and 5 mm, Schulz-Gulde and Worzyk (1983) reported experimental variations of the electric field versus current intensity. These values are shown in figure 8 and compared with our theoretical results obtained for several conditions. For a 5 mm diameter we have drawn two curves corresponding to SF_{10} calculation (with and without radiation absorption) and the curve calculated for SF_6 with absorption. For a 3.2 mm diameter the same curves have been presented, the SF_{15} conditions replacing the previous SF_{10} ones. The main comments are as follows.

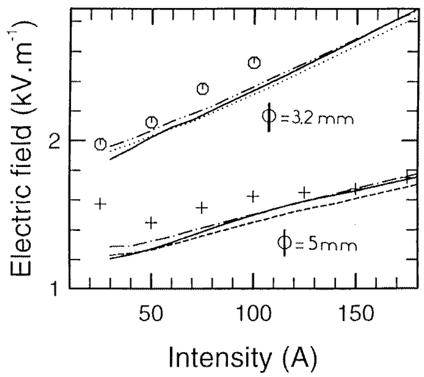

Figure 8. Variation of the electric field versus the current intensity for arc diameters of 3.2 mm and 5 mm (experimental and theoretical values). For ϕ = 3.2 mm: Schulz-Gulde and Worzyk (1983), ○; SF_6, ——; SF_{15}, ······; SF_{15} without radiation absorption, — · · —. For ϕ = 5 mm: Schulz-Gulde and Worzyk (1983), +; SF_6, ——; SF_{10}, – – –; SF_{10} without radiation absorption, — · —.

(i) Agreement between experiment and calculation is not bad but not as good as in the case of temperature studies; the experimental values are in general higher than the computed ones.

(ii) Paradoxically the best agreement is obtained when demixing is not taken into account (pure SF_6 curves).

(iii) The minimum in the characteristic curve $E(I)$ is observed experimentally at a current of 50 A ($\phi = 5$ mm) whereas this minimum occurs at a lower current in the calculation.

Four reasons may explain the differences. (i) An experimental uncertainty of 5% exists, following Schulz-Gulde and Worzyk (1983). Furthermore the measured voltage is that of the stabilizing discs and may differ systematically from the plasma voltage; but we think that this difference should be weak. (ii) The radiation absorption in the model tends to decrease the electric field (see figure 8) because the relatively warm regions ($T < 10\,000$ K) are heated by this mechanism, increasing the electrical conductance G (see equation (3)). We think that our model overestimates the absorption in this region and underestimates it in the coldest regions ($T <$ 3000 K). (Note that the radiation absorption has a very weak influence on the axis temperature; for example, in a 5 mm diameter arc the calculations show that this influence is lower than 200 K in all the current range.) (iii) From equation (3), it can be seen that the influence of the outer regions of the plasma (typically 6000 K $< T <$ 10 000 K) on the conductance is important, because of the term $r\,\mathrm{d}r$ in the integral. Some departures from equilibrium, which certainly take place in this region, may partially explain the differences between experiment and calculation for the electic field. (iv) The biggest differences ($\phi = 5$ mm and $I < 50$ A) may occur because the arc is not well stabilized when the current intensity is low and the diameter rather large.

To conclude this study about the electric field it seems that the slight disagreement between experimental and theoretical values is not due to demixing but involves other phenomena not taken into account in the experiments or in the calculation. It therefore appears that a comparison between experiment and calculation concerning the electric field does not provide a precise validation of the influence of demixing, whereas this comparison carried out on axis temperature values is a good test.

5. Conclusion

This paper has shown that demixing should be taken into account in physical models of stationary arcs established in a mixture of gases or in a multi-species gas. In the particular case of SF_6, the difference between theoretical results of axis temperature, obtained with and without considering the demixing effect is higher than the experimental uncertainty. Thus, the use of experimental results from wall-stabilized arcs for validating SF_6 plasma properties should take into account the influence of demixing on the stationary arc characteristics. Among the properties perturbed by the demixing effect, electrical conductivity and, especially, radiation losses most influence the arc characteristics.

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